

A new scintillator: Nd^{3+} doped LaBr_3

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Nd^{3+} doped single crystals, $\text{LaBr}_3:1.25\% \text{Nd}^{3+}$, $\text{LaBr}_3:2.5\% \text{Nd}^{3+}$, $\text{LaBr}_3:5\% \text{Nd}^{3+}$ and $\text{LaBr}_3:10\% \text{Nd}^{3+}$, were grown by vertical Bridgman process. The scintillation properties including light output, energy resolution, time profile, energy proportionality of the single crystals and the luminescence of polycrystalline power were measured. $2.5\% \text{Nd}^{3+}$ doped LaBr_3 exhibited the best scintillation properties while other doped ones weren't so transparent after polished.

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1. Introduction

Nd^{3+} doped LaBr_3 is a new scintillator composition that hasn't been reported before. Its scintillation properties aren't so good as previous developed scintillator $\text{LaBr}_3:\text{Ce}^{3+}$, but all the luminescence region of Nd^{3+} in $\text{LaBr}_3:\text{Nd}^{3+}$ lies on visible light. In fact, the single crystal $\text{LaBr}_3:\text{Nd}^{3+}$ appear purple, which makes it a potential scintillator matching with photomultiplier(PMT) that has a wave response of specific light region.

2. Experimental procedures

$\text{LaBr}_3:\text{Nd}^{3+}$ crystal has a hexagonal(UCl_3 type) structure with $\text{P6}_3/\text{m}$ space group, the same with $\text{LaBr}_3:\text{Ce}^{3+}$. LaBr_3 has a density of 5.1 g/cm^3 and melts at $783 \text{ }^\circ\text{C}$. NdBr_3 has a density of 5.3 g/cm^3 and melts at $684 \text{ }^\circ\text{C}$. The low melting points of these materials allow us to grow the crystals in sealed quartz ampoules by the vertical Bridgman process.

The quartz ampoules were cleaned with water, acetone and ethanol, and then baked in the oven with a temperature of $180 \text{ }^\circ\text{C}$ for 12 hours. Then the ampoules were transferred to the glove box with a humidity of less than 4%. Anhydrous LaBr_3 and NdBr_3 polycrystal were loaded into quartz ampoules in the glovebox, and then the ampoules were quickly attached to a mechanical pump and diffusion pump sequentially, evacuated to less than 10^{-3} Pa , and sealed.

The crystals were grown in a two-zone vertical Bridgman furnace (Fig. 1). A stepper motor was used to slowly lower the sealed quartz ampoule at a rate of 1.1 mm/h . The ampoules filled with molten $\text{LaBr}_3:\text{Nd}^{3+}$ moved through a temperature gradient of $30 \text{ }^\circ\text{C/cm}$ at the melting points of the mixtures.

The Hitachi F-4500 FL Spectrophotometer was used to get the fluorescence spectrum of $\text{LaBr}_3:14.2\% \text{Nd}^{3+}$ polycrystalline power. When the fluorescence spectrum was measured, the PMT voltage was 700 V , and the response time was 0.1 s .



Fig. 1. The single crystal grown furnace using vertical Bridgman method.

3. Results and discussion

In the fluorescence experiment, 350 nm wavelength was used to excite the $\text{LaBr}_3:14.2\% \text{Nd}^{3+}$ (Fig. 2).

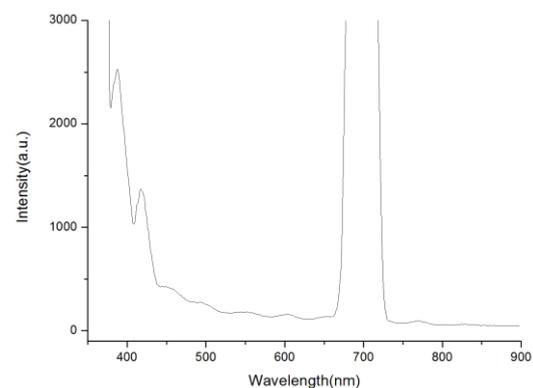


Fig. 2. The emission spectrum of polycrystalline power $\text{LaBr}_3:14.2\% \text{Nd}^{3+}$. The excitation wavelength is 350 nm .

From Fig. 2, seven fluorescence peaks due to Nd^{3+} emission are found. Due to the secondary scattering effect, the emission peaks near 700 nm may can't be observed. The assignment of Nd^{3+} emission lines is summarized in Table 1 [1].

Table 1. Emission wavelengths, wave numbers, and assignments of Nd^{3+} emission lines in $\text{LaBr}_3:14.2\% \text{Nd}^{3+}$.

$\lambda(\text{nm})$	$\nu(\text{cm}^{-1})$	Transition
387	25826	$^2\text{P}_{3/2} \rightarrow ^4\text{I}_{9/2}$
417	23969	$^2\text{D}_{5/2} \rightarrow ^4\text{I}_{9/2}$
445	22462	$^2\text{P}_{1/2} \rightarrow ^4\text{I}_{9/2}$
480	20825	$^4\text{G}_{9/2} \rightarrow ^4\text{I}_{9/2}$
551	18142	$^4\text{G}_{7/2} \rightarrow ^4\text{I}_{9/2}$
604	16551	$^4\text{G}_{5/2} \rightarrow ^4\text{I}_{9/2}$ ($^4\text{D}_{3/2} \rightarrow ^4\text{F}_{3/2}$)
769	13001	$^4\text{S}_{3/2} \rightarrow ^4\text{I}_{9/2}$

Fig. 3 shows the crucible with no-cracking single crystal after growth.



Fig. 3. The no-cracking $\text{LaBr}_3:\text{Nd}^{3+}$ single crystal grown by vertical Bridgman process. The inner diameter of quartz crucible is 25 mm.

After growth, the crystals were cut and polished with oil to minimize reaction with moisture in the air, ready for experiments (Fig. 4).



Fig. 4. $\text{LaBr}_3:2.5\% \text{Nd}^{3+}$ Crystal ready for scintillation experiments is covered with Teflon tape and packaged by Al shell and quartz glass. The inner diameter of Al shell is 17 mm.

The crystals were coupled to a Hamamatsu R6233-100 PMT while the scintillation properties except time profile were measured. The 2.5% $\text{LaBr}_3:\text{Nd}^{3+}$ crystal shows large light output while other three doped ones show really small that nearly can't be observed. The absolute light output of scintillators were measured by comparing their response with a calibrated $\text{LaBr}_3:8\% \text{Ce}^{3+}$ crystal under the same test condition.

In order to make our interesting spectrum clearer, the spectrum of the calibrated crystal is not shown. Fig. 5 is the ^{137}Cs 662 keV gamma radiation pulse height spectra measured with $\text{LaBr}_3:2.5\% \text{Nd}^{3+}$, under different shaping time: 12, 8 and 0.5 μs , and the light output is 20000 (100%), 16700 (83.5%) and 14900 (74.5%) photons/MeV, respectively.

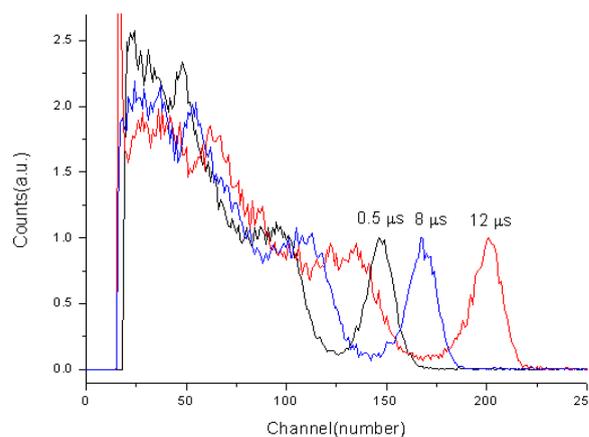


Fig. 5. ^{137}Cs source scintillation pulse height spectra measured with $\text{LaBr}_3:2.5\% \text{Nd}^{3+}$ under different shaping time: 12, 8, and 0.5 μs .

The crystals were coupled to a Hamamatsu XP2020Q PMT while time profile was measured.

Fig. 6 is the time profile of $\text{LaBr}_3:\text{Nd}^{3+}$. The principal decay time of scintillator is 15 ns for each of the four different Nd^{3+} concentrations.

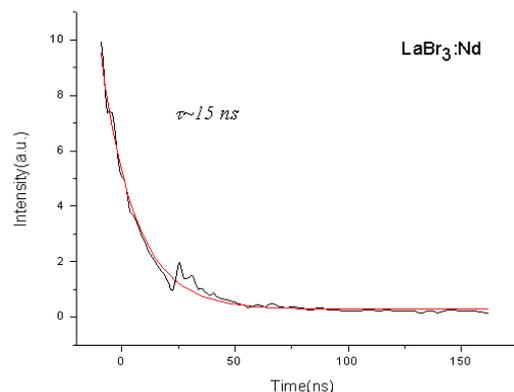


Fig. 6. Decay time of $\text{LaBr}_3:\text{Nd}^{3+}$.

Among the four concentrations doped LaBr₃, 1.25% Nd³⁺, 2.5% Nd³⁺, 5% Nd³⁺ and 10% Nd³⁺, it's found that the 2.5% Nd³⁺ doped LaBr₃ is the most transparent after polished.

Table 2. The scintillation properties of LaBr₃:2.5%Nd³⁺.

Light Output (12 μs, photons/MeV)	Energy Resolution (@662 keV)	Principal Decay Time (ns)	Nonproportionality (60~1200 keV)
20000	7.5%	15	27%

The proportionality of LaBr₃:2.5% Nd³⁺ was evaluated by measuring their responses to gamma ray of ²⁴¹Am (59.5 keV), ¹³⁷Cs (662 keV), ¹⁵²Eu (121.78, 244.69, 344.28 keV) and ⁶⁰Co (1170 keV) (Fig. 7). The data were normalized with respect to the measured light output at 662 keV.

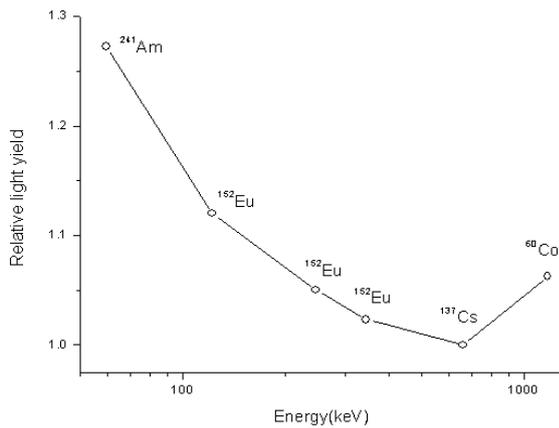


Fig. 7. Relative light yield versus energy for LaBr₃:2.5%Nd³⁺.

The information of light output, energy resolution, principal decay time and nonproportionality of LaBr₃:2.5% Nd³⁺ are listed in Table 2.

4. Conclusions

The luminescence of polycrystal and scintillation properties of Nd³⁺ doped LaBr₃ single crystals were measured. The LaBr₃:2.5% Nd³⁺ exhibits the best scintillation properties, with the light output, energy resolution, principal decay time and nonproportionality to be 20000 photons/MeV, 7.5%, 15 ns, 27%, respectively.

Acknowledgements

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References

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